



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re: Patent Application of : Group Art Unit: 1754
Iwao ANZAI et al.

Serial No.: 10/813,473 : Examiner: WAYNE A. LANGE

Filed: March 30, 2004

For: Process of : Attorney Docket No.:
Producing Fuel 208305.0241/223U1
Gas for Fuel Cell (NP136-1D)

DECLARATION UNDER 37 C. F. R. § 1.132

I, Iwao Anzai, declare and state as follows:

1. I am a graduate of the Faculty of Engineering at Kyoto University in March 1984.

2. In April 1984, I was employed by Nippon Oil Co., Ltd. Ltd. Currently, I am employed by Nippon Oil Corporation in the Central Technical Research Laboratory, where I have been

actively engaged in the research and development of catalysts, focusing on particularly autothermal reforming catalysts.

3. I am the same Iwao Anzai who is a co-inventor and co-applicant in the above-identified patent application. I am at least a co-inventor of all of the presently pending claims and conducted experiments described below.

4. I have reviewed the Office Action dated October 29, 2004 in the above-identified application, and studied WO99/64150, U.S. Patent Application Publication No. 2004/0014600 (Fukunaga '600), U.S. Patent Application Publication No. 2002/0150532 (Grieve), U.S. Patent Application Publication No. 2002/0009408 (Wieland), and U. S. Patent No. 5,268,346 ("Ino"), which the Examiner has used to reject all of the pending claims under 35 U. S. C. § 103 (a).

5. It is my understanding that the Examiner is of the position that claims 1 to 4 are obvious over WO99/64150, Fukunaga '600, Grieve et al., or Wieland et al., all of which disclose autothermal processes for producing a fuel gas wherein the catalyst comprises ruthenium on an alumina support, wherein the catalyst may also contain cerium oxide, in view of Ino et al. which discloses a catalyst for steam reforming of hydrocarbons comprising a platinum group metal on a carrier which comprises 5 to 40 weight percent of ceria and 60 to 95 weight percent of alumina. It is also my understanding that the Examiner was relied on the disclosures of Wieland et al. and Grieve et al. because both of them suggest the equivalence between steam reforming and autothermal reforming. However, all catalysts suitable for steam reforming are not necessarily

suitable for autothermal reforming. Therefore, it would not be obvious for a person skilled in the art to employ the carrier of Ino et al. for steam reforming as the carrier for the catalyst in the process of either Wieland et al. or Grieve et al.

6. This Declaration is submitted to prove that catalysts exhibiting excellent properties in steam reforming do not necessarily exhibit excellent properties in autothermal reforming. I have conducted additional comparative experiments employing Catalysts I and J used in the Examples described in the specification and Catalyst K which was newly prepared for the experiments. More specifically, I conducted additional experiments wherein these Catalysts I to K were used for steam reforming and also Catalyst K was used for autothermal reforming.

7. Catalyst K was prepared as follows:

Preparation of Catalyst K:

γ -alumina powder with a specific surface area of $190 \text{ m}^2/\text{g}$ was calcined in the air at a temperature of 800°C for 3 hours and then dipped into a water-soluble nickel solution. After the water was evaporated, the powder was dried at a temperature of 120°C for 3 hours. After the powder was pressed, it was ground and shifted, thereby obtaining a granulated catalyst with a size of about 1 to 2 mm. The catalyst was reduced under a hydrogen circulation at a temperature of 500°C for 3 hours thereby obtaining Catalyst K. The chemical composition of Catalyst K is set forth in Table 1 below.

8. Steam reforming reaction was conducted as follows:

Steam reforming reaction

A reaction tube with an inner diameter of 9 mm was one cc of Catalyst K and then set in a tube-like electric oven. A steam reforming reaction was conducted by introducing desulfurized kerosene as the feed stock at an LHSV of 2.0 h⁻¹ into the reaction tube at a reaction temperature of 700 °C, at a steam/carbon ratio (molar ratio) of 3.0, and at atmospheric pressure, for 20 hours.

The reaction gas was analyzed using a gas chromatogram to measure the amount of each H₂, CO, CO₂, and CH₄. The conversion rate was determined by calculating the percentage of CO, CO₂, and CH₄ converted from the feed stock. The results after 20 hours are set forth in Table 1.

The same steam reforming reaction was repeated by using Catalysts I and J. The results are also set forth in Table 1.

9. Autothermal reforming reaction was conducted as follows:

Autothermal reforming reaction

An autothermal reforming reaction was conducted in the same manner as the Examples described in the specification of the instant application using Catalyst K. The conversion rate and composition of the reformed gas after 20 hours are set forth in Table 2.

Table 1

Feed Stock	Desulfurized Kerosene	Desulfurized Kerosene	Desulfurized Kerosene
Catalyst	I	J	K
Catalyst Composition (mass%)			
Al ₂ O ₃	99	balance	80
CeO ₂	-	1	-
Ba	-	15	-
Rh	1	1	-
Ni	-	-	20
Conversion Rate (%)	100	100	100
Carbon Precipitation Amount (mass%)	0.9	0.8	4
Composition of Reformed Gas (volume%, dry basis)			
H ₂	71.2	71.3	71.0
CO	15.1	15.0	15.1
CO ₂	12.9	12.8	13.0
CH ₄	0.8	0.9	0.9

Table 2

Feed Stock	Desulfurized Kerosene	Desulfurized Kerosene	Desulfurized Kerosene
Catalyst	I	J	K
Catalyst Composition (mass%)			
Al ₂ O ₃	99	balance	80
CeO ₂	-	1	-
Ba	-	15	-
Rh	1	1	-
Ni	-	-	20
Conversion Rate (%)	89	91	71
Carbon Precipitation Amount (mass%)	1	0.8	4.8
Composition of Reformed Gas (volume%, dry basis)			
H ₂	46.1	46.0	43.5
CO	6.4	6.3	5.1
CO ₂	16.1	16.0	17.5
CH ₄	0.3	0.3	1.2
O ₂	0.2	0.3	0.2
N ₂	30.9	31.1	32.5

10. As apparent from Tables 1 and 2, Catalyst K was high in conversion rate in the steam reforming reaction. However,

in the autothermal reaction, Catalyst K was lower in conversion rate even than Catalysts I and J which were used as Comparative Examples for the present invention. Furthermore, Catalyst K was larger in the amount of carbon precipitation than Catalysts I and J.

11. I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: 04-21-2005

Iwao Anzai

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